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OAK RIDGE NATIONAL LABORATORY
Operated by
UNION CARBIDE NUCLEAR COMPANY
Division of Union Carbide Corporation
Post Office Box X
Oak Ridge, Tennessee

DATE: January 10, 1961

COPY NO. 59

SUBJECT: Plutonium Release Incident at Oak Ridge National Laboratory*

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ABSTRACT

A nonnuclear explosion involving an evaporator occurred in a shielded cell in the Radiochemical Processing Pilot Plant at Oak Ridge National Laboratory. Plutonium released from the processing cell contaminated areas in the pilot plant building and nearby streets and building surfaces. The explosion is considered the result of rapid reaction of nitrated organic compounds formed by the inadvertent nitration of about 14 liters of a proprietary decontaminating reagent.

In cleanup the contamination was bonded to the nearby street and building surfaces with tar, paint, roofing compound, or masonry sealer, as appropriate to the surface. Decontamination of the interior of the pilot plant building, except the processing cells, was 95% complete on September 1, 1960.

*Presented at the ANS Winter Meeting, San Francisco, California, December 12-14, 1960.

On November 20, 1959, a nonnuclear explosion involving an evaporator occurred in a shielded cell in the Radiochemical Processing Pilot Plant at Oak Ridge National Laboratory. Plutonium released from the processing cell contaminated portions of the pilot plant building and nearby streets and building surfaces. No radioactivity was released from ORNL. No one was injured by the explosion. No one received a significant fraction of a lifetime body burden of plutonium or an overexposure to sources of ionizing radiation either at the time of the incident or during subsequent cleanup operations.

The explosion was probably the result of rapid reaction of nitrated organic compounds formed by the inadvertent nitration of 14 liters of Turco Decon 4501, a proprietary decontaminating agent which is a strongly basic solution of the alkali salts of various organic hydroxy acids, various amines, surface active agents, and phenol.

The cell in which the explosion occurred is one of four cells containing equipment for solvent extraction processing of highly irradiated nuclear fuels. About one month preceding the incident, after several processing campaigns with highly irradiated uranium had been completed, a decontamination program was begun. One objective of this program was to decontaminate the evaporator subcell which was emanating greater than 100 r/hr of penetrating radiation.

The decontamination sequence as normally used in the pilot plant consisted of a Turco Decon 4501 treatment at 180-190°F, a water flush, a 30% HNO_3 treatment at 180-190°F and a final water flush. The intercycle evaporator was treated with one complete sequence, with a hot 25% NaOH flush and with extensive water rinses, but no reduction in radiation levels occurred.

It was decided to make another Turco Decon 4501, water, and acid treatment. Turco Decon 4501 was added to the evaporator and heated for 2 hr.

The evaporator drain valve was located in a high level radiation field (the one responsible for the decontamination program). For this reason the Turco Decon 4501 solution was drained from the evaporator via the product outlet valve and a 16.5 liter heel was left which contained about 14 liters of "as received" Turco Decon 4501. Inadvertently, the

water flush was omitted and 30% HNO_3 was added to the evaporator where it mixed with the organic decontaminating agents.

Normally the acid would be heated just to 180-190°F but this particular time it was boiled. This provided sufficient heat to concentrate the nitric acid and allow complete reaction between the acid and organic compounds. As the acid became concentrated the temperature increased steadily until at about 230°F the nitro compounds, just formed, ignited and exploded.

The sequence of events which led to the explosion and which were necessary for the explosion are:

1. Inadvertent omission of water rinse.
2. Incomplete draining of the Turco Decon 4501 from the evaporator.
3. Boiling the nitric acid.

Physical damage from the explosion was confined to the intercycle evaporator equipment and its unit shield wall of dry stacked barytes concrete blocks located in one corner of the cell.

Figure 1 is an isometric drawing of the major evaporator components before and after the explosion. The subcell is 5 ft wide by 7-1/2 ft deep by 15 ft high. The vapor separator (P-2) was torn apart by the indident. A large, badly deformed section that originally was part of the side and top and bottom was found lying on the floor in the subcell. The remainder of separator P-2 was found wrapped around the steam stripper (P-5) which was dented by the impact.

The convection loop (P-15 and P-7) was completely separated from the vapor separator. The flared end of the heat exchanger (P-7) nozzle failed circumferentially and the 2-in.-dia pipe which had connected the heat exchanger (P-7) to the vapor separator (P-2) was found lying on the floor.

Figure 2 is a photograph of the equipment after the explosion.

The three chemical operators and foreman were in the control room when they heard the explosion and felt the building shake. They saw the make-up room filling with a fog and they evacuated the building. The foreman and one operator put on gas masks and went back into the building to make a quick check for radiation and to secure the plant.

Operating, Health Physics and management personnel were called to the plant and access to the area was restricted. A plan was initiated to

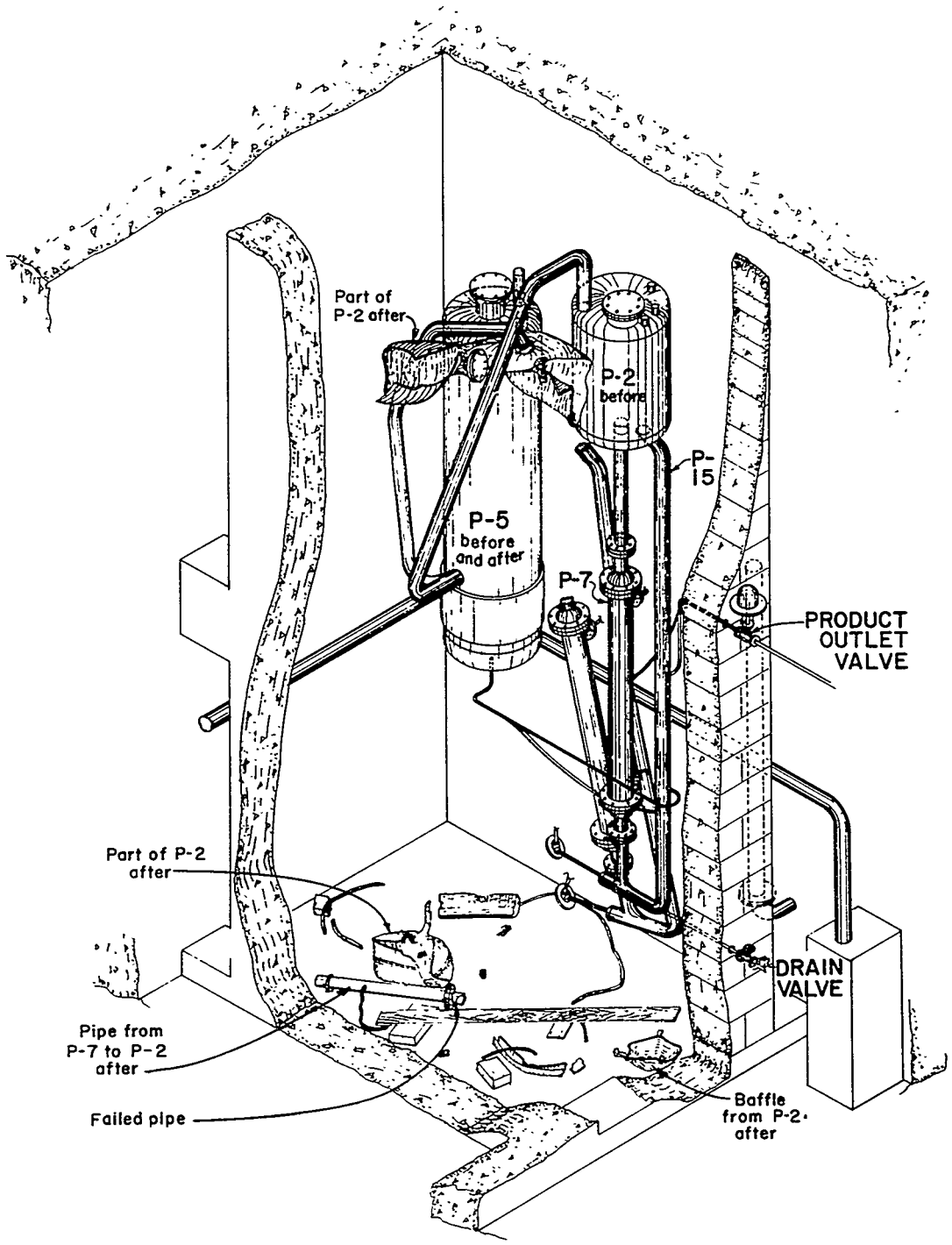

Fig. 1. Intercycle evaporator before and after explosion.

Fig. 2. Intercycle evaporator after explosion.

evaluate the situation and start cleanup operations.

The momentary increase in cell air pressure which must have accompanied the explosion caused air-borne activity to be transported from the processing cell to other areas in the building and to the outside of the building.

Figure 3 is a map of the Laboratory which shows the Radiochemical Processing Pilot Plant and the portion of the Laboratory which was involved in the fall-out. Figure 4 is an enlargement of the affected area. Contamination in some areas was more than 100,000 disintegrations per minute per 100 sq cm. This is about one bpdy burden of plutonium per 100 sq cm.

Fortunately the fall-out zone was small because there was only a slight breeze blowing and fall-out was rapid. A Health Physics surveyor drove from the Oak Ridge Research Reactor to the cell 6 door, got out of his truck, and looked toward the cell 6 door from the road; all within 2 minutes after the explosion. The truck was completely contaminated but the surveyor only got his shoes contaminated.

The filters in the cell off-gas system retained the alpha activity with a high degree of efficiency. The filter bank is such that the off-gas passed through two layers (FG-25 filterdown and FG-50 filterdown) as a roughing filter and a high efficiency or "absolute" filter. Of a calculated total of 1500 mg of plutonium retained on the filters, 1325 mg was retained on the first layer of roughing filter, 156 mg on the second layer and 18 mg on the absolute filter. It is felt that no plutonium penetrated to the back side of the absolute filter.

The interior of the Graphite Reactor Building (3001) was contaminated by plutonium which was drawn into and through the building by the ventilation system. Contamination found in other buildings was slight and was readily removed by ordinary cleaning methods.

Containment of the radioactivity was the first consideration. A general wash down of the area was not attempted; first, because contamination might be washed into crevices where it would be inaccessible for cleanup but would be present as a contributor to high air activity levels, and second, because the amount of water which could be held in the waste storage basins was limited. It was decided to take steps to "fix" the activity until it could be removed in an orderly way at some later time. Badly contaminated roads and the roof of Building 3025, Solids State

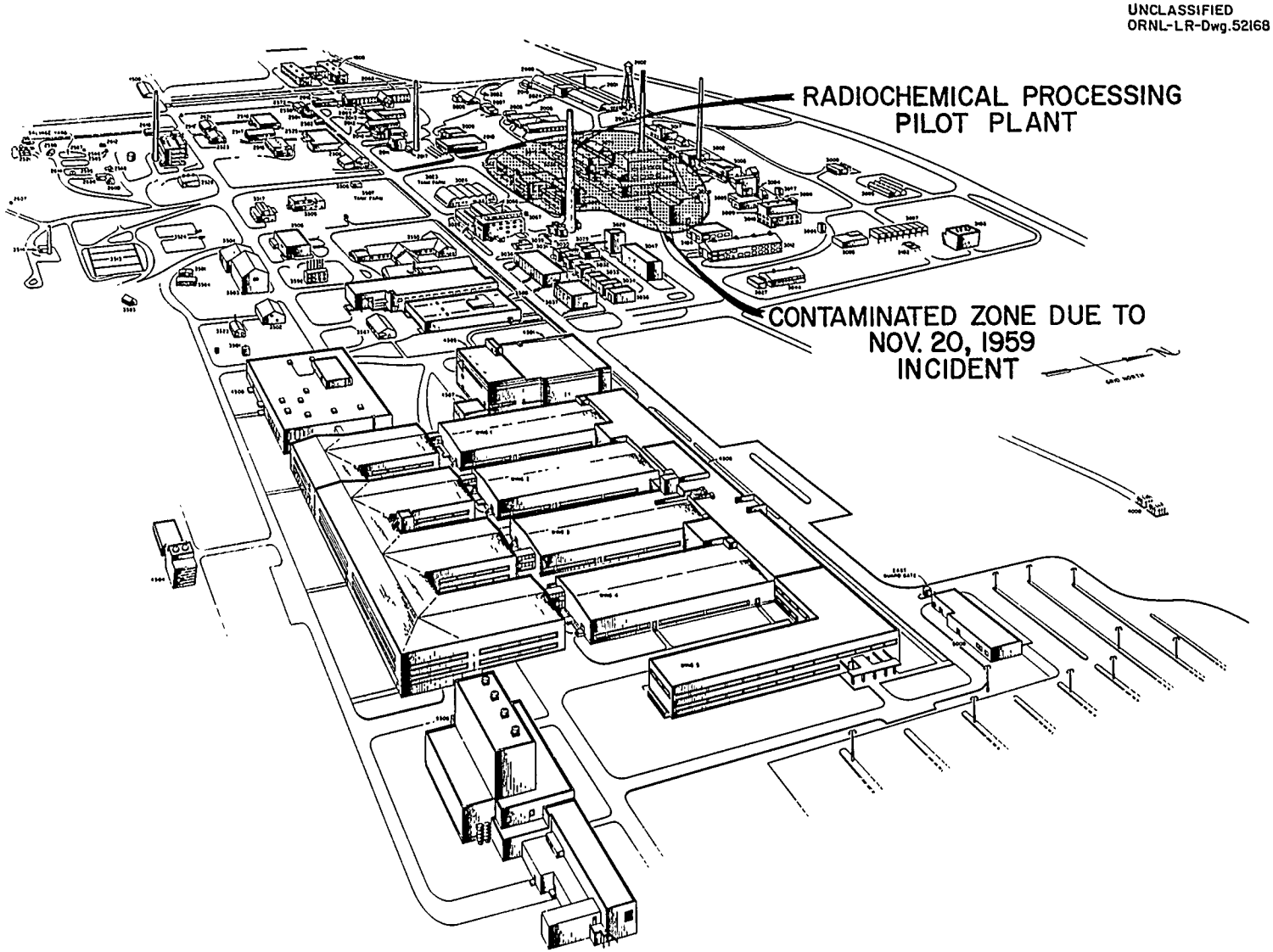

Fig. 3. Oak Ridge National Laboratory,
X-10 Site

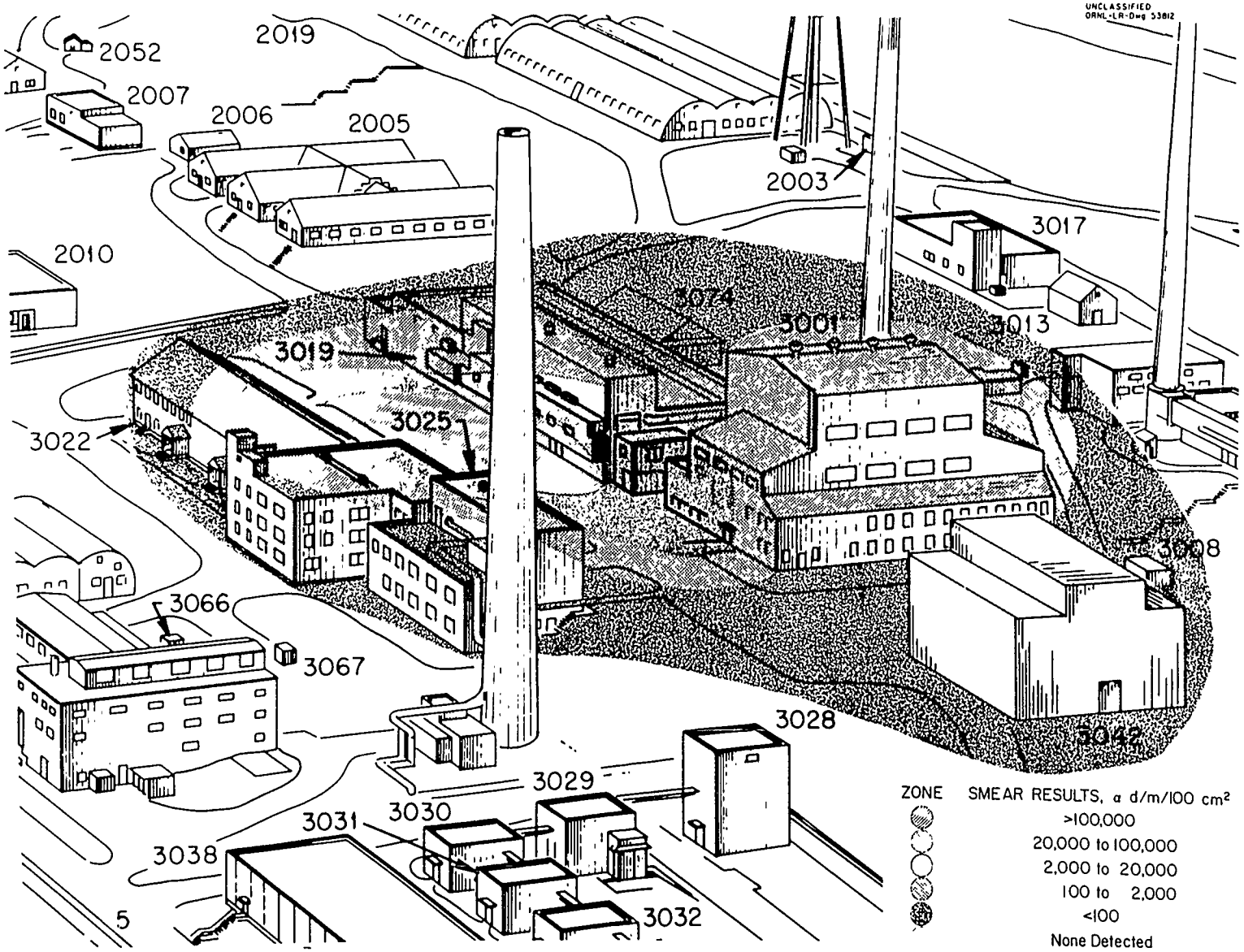

Fig. 4. Plutonium fallout after explosion

Physics, were resurfaced with emulsified asphalt and gravel. Paint was sprayed on everything else in the area - including roofs, walls, grass, pipes, barrels, and sidewalks. The interiors of nearby buildings were surveyed and cleaned as needed. Except for the Pilot Plant portions of Building 3019, the Graphite Reactor Building, and the roadway just south of these buildings, all Laboratory areas were back in service on Monday morning, November 23.

Figure 5 is a sectional elevation of Building 3019 through the evaporator. The plutonium alpha activity which contaminated streets and buildings in the vicinity of the Radiochemical Processing Pilot Plant was released from that building through the cell 6 door, which is at ground level and opened directly to the outside of the building.

The contamination in the building ranged from 10^8 alpha d/m/100 cm^2 in the cell to 50 disintegrations per minute per 100 cm^2 in the offices. Outside the cells, the greatest contamination in the building was in the penthouse.

Open access holes from the processing cell to the make-up area, pipe tunnel, sampling gallery, and penthouse were the paths by which the plutonium escaped the cells to contaminate the interior of Building 3019.

Of the 3500 man days of cleanup about one third were expended in the penthouse. The most difficult area was the 167 by 30 ft ceiling which is constructed of ribbed metal roof decking supported by I-beams. After the unsuccessful use of scrubbing with detergents, scraping, and sanding, the only effective decontamination method found was complete removal of all paint with a chemical paint remover.

Floors were cleaned by removal of the concrete surface and were covered by tile or new concrete and paint. Except for stainless steel which responded to strong acid cleaning, unpainted metal surfaces were particularly difficult to decontaminate and often required grinding until the surface was brightly polished before cleaning specifications were reached.

Cleanup in cell 6 was delayed 8 months until building modifications could be made to improve containment. Cell equipment was flushed using a solution of 2 M HNO_3 and 0.02 M fluoride, with 0.04 M boron added as a neutron poison.

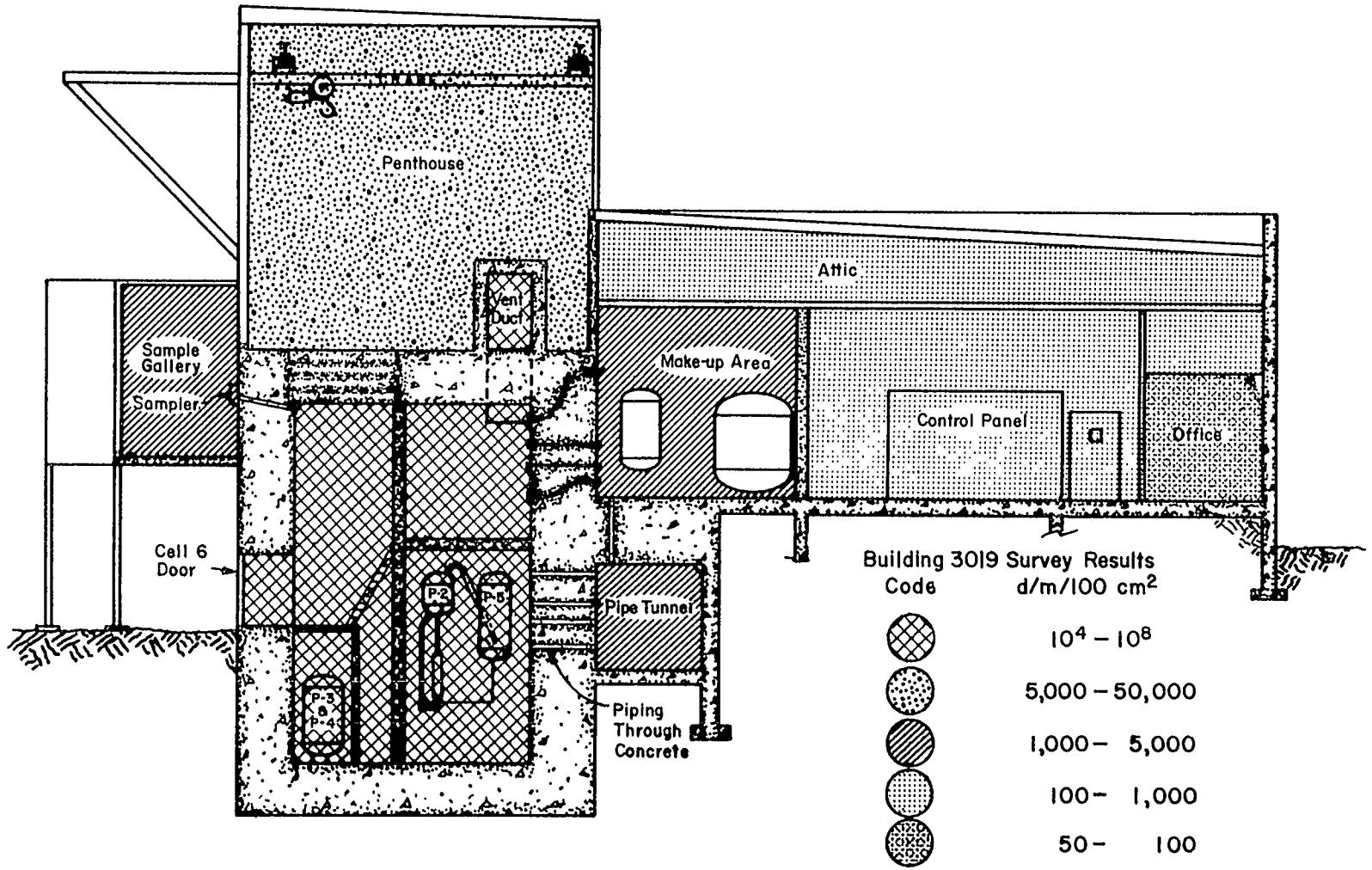

Fig. 5. Sectional elevation through cell 6, Radiochemical Processing Pilot Plant, showing inside contamination levels after explosion.

About 700 g of plutonium was flushed from the ruptured evaporator (mostly P-5). About 400 g was flushed from product catch tank P-3 and this also is believed to have come from the ruptured evaporator. About 10 g was found in the rest of the process equipment.

There are two strip column-evaporator combinations which were operated in parallel during most of 1959. The evaporator not involved in the explosion had been decontaminated and repaired prior to the incident but no plutonium was found. During a short processing campaign which used only this "other" evaporator the plutonium material balance showed a "profit" of ~170 g. Only 3 g was flushed from this evaporator during the latest equipment flush.

There is no plausible explanation of why plutonium collected preferentially in steam stripper P-5.

Much of the debris has been removed from the cell and cell surfaces have been hosed down. Washings contained about 120 g of plutonium. The level of penetrating radiation in the cell has been reduced to a general background of about 50 mr/hr with places as high as 2 r/hr.

The alpha contamination is still high enough that personnel are required to wear plastic suits to enter the cell but final cleanup of the cell will be a straightforward operation.

Following this incident all high-level radiochemical operations at ORNL were suspended until all the hazards could be re-evaluated. Radioactive operations at ORNL (and these include radiochemical pilot plants, "hot" labs, and experimental set-ups, as well as ORNL reactors) must satisfy the following general criteria:

1. The maximum credible accident must be contained or confined to the degree necessary to preclude discharge into the Laboratory area and its environment of concentrations or amounts of radioactive materials which are injurious to health or which will interfere with other Laboratory programs.
2. Two lines of defense must be present to prevent the escape of radioactive materials from Laboratory operations via waste streams.

These criteria apply not only to future operations, but also to those in existence. A formal hazards report is required for those operations involving >1 g of plutonium (or equivalent of other alpha emitters or isotopes

of comparable hazards) or >1000 curies of beta and gamma emitters. The reports will be submitted to the appropriate ORNL review committee and must be approved by the committee and ORNL Management prior to initiation or resumption of operation.

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